ISSN 1066-3622, Radiochemistry, 2017, Vol. 59, No. 4, pp. 365–371. © Pleiades Publishing, Inc., 2017.
Original Russian Text © I.V. Smirnov, E.S. Stepanova, M.Yu. Tyupina, N.M. Ivenskaya, I.G. Tananaev, S.R. Zaripov, S.R. Kleshnina, S.E. Solov'eva, I.S. Antipin, 2017, published in Radiokhimiya, 2017, Vol. 59, No. 4, pp. 319–324.

Effect of Ionizing Radiation on the Extraction of Am(III) with *p-tert*-Butylthiacalix[4]arene from Alkaline Carbonate Solutions

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Received December 12, 2016

Abstract—The effect of γ-irradiation of *tert*-butylthiacalix[4]arene (TCA) solutions in *m*-nitrobenzotrifluoride (NBTF) and tetrachloroethylene (TCE) on the extraction of ^{241}Am from alkaline carbonate solutions was studied. TCA itself remains stable upon γ-irradiation of its solutions in NBTF to a dose of 200 kGy, but the diluent undergoes strong degradation. The radiation resistance of TCA in TCE is considerably lower: A dose of 70 kGy causes complete degradation of TCA. In the TCA–TCE–aqueous phase system, sulfate ions appear upon γ-irradiation as the final product of the extractant radiolysis. A large number of γ-radiolysis products of TCE and TCA were detected by HPLC and GCMS. The products of radiolysis of TCA in TCE, compared to the initial extractant, have lower molecular mass and higher polarity. The results show that chlorinated diluents are not promising diluents for thiacalixarene in extraction processing of alkaline high-level waste.

Keywords: extraction, americium-241, thiaxalix[4]arene, alkaline media, γ-radiolysis, radiolysis products

DOI: 10.1134/S1066362217040087

One of the most important problems of modern applied radiochemistry is final liquidation of so-called "nuclear heritage." Active efforts are made at nuclear objects for remediation of contaminated territories, decommissioning of large nuclear facilities, and final removal and disposal of the accumulated radioactive waste. One of the aspects of this problem is processing of high-level waste (HLW) accumulated in the course of implementation of the USSR Nuclear Project at the Mayak Production Association. This HLW has a volume of more than 18000 m^3. It is in the form of multicomponent slurries containing sodium hydroxide, carbonate, nitrite, and nitrate in combination with such hazardous radionuclides as ^{137}Cs, ^{90}Sr, and transuranium nuclides [1–3].

The first step of processing of complex multicomponent HLW should involve recovery of hazardous radionuclides from the liquid phase of the heterogeneous medium by one or another procedure. This study was aimed at developing a process for extraction preconcentration and separation of ^{241,243}Am from multicomponent alkaline media using high-performance macrocyclic compounds. We have shown that Am(III) is efficiently extracted from alkaline carbonate solutions with substituted thiacalix[4]arenes. *p*-Bromothiacalix[4]arene at pH 12 showed the highest ability to extract Am (distribution ratio $D_{Am} > 100$) and the highest selectivity [selectivity factor β_{max}(Am/Eu) = 18] [4].

The suggested promising extraction system for recovering radionuclides from alkaline carbonate media should be adapted to real conditions by performing